

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

**Applicant(s): Donald R. Huffman, et al.**

**Examiner: Hendrickson**

**Serial No.: 07/580,246**

**Art Unit: 1754**

**Filed: September, 10th 1990**

**Docket: 7913Z**

**For: NEW FORM OF CARBON**

**Assistant Commissioner for Patents  
Washington, DC 20231**

**DECLARATION OF Raouf O. Loutfy**

I, Raouf Loutfy, hereby declare:

1. I reside at 6507 N. Ventana Canyon Drive, Tucson, Arizona, USA.
2. I earned a B.Sc. degree in 1964 in Applied Chemistry from Cairo University, a M. Sc. in 1966 in Solid State Sciences from the American University, a Ph.D. in 1971 in Physical Chemistry from University of Western Ontario, and a Diploma in Business Administration in 1976 from McGill University.
3. Between 1977 and 1981 I was employed as a group leader at Argonne National Laboratory in the Chemical Engineering Division.

4. Between 1981 and 1985 I was employed as Research Advisor of ARCO Metals/ARCO Chemical developing advanced material technologies.

5. Between 1985 to present I have been employed as president of MER Corporation, and since 1990 I have been involved in the commercial scale-up of fullerenes, and in developing applications for fullerenes.

6. I have published many articles, two books, and contributed a chapter in Encyclopedia of Technology on the fullerene production, and on applications of fullerenes. I have been an invited speaker to many technical and investment conferences as an expert in the technology and commercialization of fullerenes. I have received the prestigious Tibbetts award in 2001 from the Small Business Administration (SBA) for the commercialization of fullerenes.

For the convenience of the patent and trademark office, I have attached hereto as Exhibit 1 my curriculum vitae, which describes my credentials and demonstrates my expertise in the area of fullerenes.

7. I am intimately familiar with the published literature concerning fullerenes and I am personally involved in the research and development of new methods of production and applications for fullerenes including C<sub>60</sub>, C<sub>70</sub> and nanotubes.

8. I am informed by Mark J. Cohen, Esq., the attorney handling the prosecution of the subject United States patent application that a question has arisen concerning the use

of the term "macroscopic amounts" as applied to the fullerene products, e.g. C<sub>60</sub> and C<sub>70</sub> that are produced by the methods invented by Drs. Huffman and Kratschmer as described in the underlying application.

9. As set forth above, I have been professionally involved in the field of fullerenes development and manufacture since Dr. Huffman and Kratschmer disclosed their novel process for producing macroscopic quantities, i.e., visible quantities of fullerenes, e.g., C<sub>60</sub> and C<sub>70</sub>, as disclosed in the subject patent application and in their publication in Nature (1990) identified elsewhere in this Declaration and I am familiar with the use of the term "macroscopic quantities" in this field and, in general, in the scientific and academic communities.

10. I have read and reviewed the subject patent application, including the pending claims. It is my understanding that the subject application discloses, among other things, a process of producing fullerenes including C<sub>60</sub> and C<sub>70</sub> in macroscopic amounts and that, inter alia, U.S. Serial No. 07/580,246 includes claims directed to macroscopic quantities of C<sub>60</sub> and C<sub>70</sub>.

11. Although the subject patent application of Dr. Kratschmer and Huffman does not expressly use the term "macroscopic amounts" to describe the amounts of fullerenes, e.g., C<sub>60</sub> and C<sub>70</sub> first isolated by them, in accordance with the teaching of the process described therein, the fullerenes, e.g., C<sub>60</sub> and C<sub>70</sub>, that were prepared in accordance with the process described herein, were produced in measurable amounts that were visible to

them, and it is my professional opinion that these amounts are inherently amounts definable by the term "macroscopic amounts".

12. Part of our research and development at MER involves the use of various arc processes, plasma processes, and high temperature process to produce fine metal, ceramic, and carbon powders. In the course of this research and development work, MER has since from about 1985 to the present maintained equipment and capabilities that could have produced soot. I am quite familiar with the principal of operations and the use of such equipment. At MER, I regularly produced soot under arc, plasma, and high temperature conditions for various applications. I am also familiar with the work of the Nobel Laureate Sir Harry Kroto et al. al. published in Nature volume 318, No. 6042, 14-20 November 1985. p162. In this publication they report the detection of remarkably stable cluster of 60 carbon "atom" using time-of-flight mass spectrometry in the vapor phase. However, they never isolated or recovered visible particle of  $C_{60}$  and  $C_{70}$ , and did not disclose a process that would teach or lead others to do so. It is noted they reported the following "If a large-scale synthetic route to this  $C_{60}$  species can be found, the chemical and practical value of the substance may prove extremely high".

13. On or about September 18, 1990, MER received and executed confidentiality agreement from RCT regarding disclosure to MER of Huffman's et al. al. discoveries.

14. On or about September 20, 1990, Dr. Huffman of U of A and Mr. Jacobs of RCT visited MER Corporation to further discuss proactive strategy concerning an arrangement

to license from RCT under the Huffman et. al. invention to produce fullerenes at MER and to capitalize on the fullerene discovery of Dr. Huffman et al., and to discuss in details the production and the separation of fullerenes from the vaporized carbon utilizing the Huffman et al. discoveries.

15. On October 9, 1990, Dr. Withers, CEO of MER, and I visited Dr. Huffman's laboratory and observed his apparatus and witnessed the operation of his apparatus for vaporization of carbon, and production and collection of soot containing fullerenes. In many respects, Dr. Huffman's apparatus was very similar to much of the equipment at MER; however, MER had not used their equipment to produce fullerenes prior to the Huffman et. al. invention. The operating conditions for such equipment and the realization of the existence of fullerenes in the soot were the missing elements, not only from MER's viewpoint, but also from that of all other researchers prior to the Huffman et. al. invention. The equipment of MER and others will not produce inherently fullerene and even if produced it was not known that it could be recovered as it was later taught by Dr. Kratschmer and Huffman. All researchers in this field either produced soot under high vacuum conditions, without sufficient concentration of carbon vapor, at low pressure, or without sufficient cooling or combination of these conditions to produce recoverable fullerenes in the soot. These conditions resulted in product that does not contain fullerenes. Even though it appears simple to the uninformed, especially in hindsight, the process of Dr. Kratschmer and Huffman as described in the subject application, is a remarkable discovery, which produced a high density of vapor of carbon as described on page 4 of the subject application which resulted in the formation of

macroscopic amounts of fullerenes by their method. From 1985, when Dr. Smally et. al. at Rice University discovered the existence of  $C_{60}$  and  $C_{70}$  atoms by spectrographic analysis of a vapor (see paragraph 12 above), until Dr. Huffman et. al. published their discovery in 1990 no one else realized how to produce and recover macroscopic quantities of these fullerenes, despite the availability of equipment that could have been used for this purpose.

16. I directed experiments to produce fullerene soot at MER using equipment at MER, and we also built additional low vacuum equipment based on the Kratschmer and Huffman method described in the subject application and produced macroscopic quantities of soot and fullerene. We separated the fullerene according to their teaching. From a similar apparatus as that described in the subject application, but operated for sufficient time, we were able to offer for sale to the research community fullerenes including pure (90%+)  $C_{60}$  and  $C_{70}$  by November 1990.

We have since developed computer control, fully automated reactor equipment using Kratschmer and Huffman's teaching in the subject application and we optimized the process to maximize the yield of fullerenes.

17. I repeated exactly the Huffman et. al. process according to the teaching described in the subject application including example 1 using  $\frac{1}{4}$  inch in diameter graphite rod, at 100 torr Helium, using 100 ampere dc current. This graphite vaporized, and the vapor was condensed on a water cooled surface. The vaporization was performed for 50 minutes using about 17 cm length of the graphite rod and produced 12 gram of soot. The

fullerenes were recovered using toluene and the amount of fullerene was determined. The yield of fullerene was about 8 to 10%. Accordingly, the total recoverable fullerenes was over 1.2 grams with over 900 mg of C<sub>60</sub> and over 200 mg of C<sub>70</sub> and the remaining other fullerenes.

According to the teaching in the subject application where they vaporized a 1/4" graphite rod with 1 cm length, the inventor must have produced at least about 600 mg of soot that contains admixture of at least 63 mg of fullerenes that contain at least about 50 mg of C<sub>60</sub> and at least about 10 mg of C<sub>70</sub>. A 600-mg quantity of soot certainly can be seen by the naked eye, as also indicated by the inventor that "heavy black coating on collecting substrates and/or on the walls of the chamber which can be easily scraped off for the recovery step." Also, the 60 mg of fullerene certainly can be seen by the naked eye and it is measurable. Furthermore, the 45 and 10 mg of C<sub>60</sub> and C<sub>70</sub> respectively are also measurable, in today modern laboratory facility amount as low as 0.1 mg can be measured, and can be seen by the naked eye.

The same conclusion can be reached by simply calculating the mass of the rod vaporized in Kratschmer et al. subject application, including example 1, which is easily determined from the diameter of the graphite rod they used (1/4"), the length (1 cm), and typical density of the type of graphite used for graphite vaporization (2.0 g/cc). This calculation estimates that about 633 mg of soot containing fullerenes was produced by Kratschmer et al., which is certainly macroscopic and in agreement with the above-presented experimental data.

Moreover, if a longer graphite rod were used, the amount of C<sub>60</sub>, C<sub>70</sub> and other fullerenes produced would even be greater, as shown herein above.

18. It is in my opinion that the inventors of this subject application were the first to isolate and recover a measurable or macroscopic amount of fullerenes, and to teach others to do so. Their description in the application is clearly understood by ordinary skilled artisans, and when repeated by us allowed us to produce visible, measurable commercial quantities of fullerene product, commonly described as "macroscopic quantities".

19. It is also in my opinion that the inventors produced "macroscopic amounts" as used in the claims as clearly understood by the ordinary skilled artisan, and supported by the above. The term is commonly used to connote that amount which can be seen easily with the naked eye. This opinion is corroborated by the usage of this term by others in the fullerene field to describe the fullerene product produced by the process invented by Drs. Kratschmer and Huffman, for example, see the statement appearing at Column 1, lines 58-61 in U.S. Patent No. 6,077,401 issued June 20, 2000, on an application filed August 15, 1994, which is as follows:

"All of these applications have been discovered since the first macroscopic amounts of the most common fullerene, C<sub>60</sub>, were isolated in 1990 [Kratschmer, et al., Nature 347, 354 (1990)]."



The cited quote from the Fields et. al. patent also corroborates the common understanding of those familiar with the fullerene art that they were the first persons to produce fullerenes in macroscopic quantities and to show others how to do so.

For the convenience of the patent and trademark office, I have attached hereto as Exhibit 2 the cited patent.

I further declare that all statements made herein of my own knowledge are true, and all statements made on information and belief are believed to be true. I acknowledge that willful false statement and the like are punishable by fine or imprisonment or both (18 U.S.C. §1001) and may jeopardize the validity of the application or any patent issuing thereon.

July 16, 2002  
Date

Raouf O. Loutfy  
Raouf O. Loutfy, Ph.D

Signed in Tucson, Arizona

## **Exhibit # 1**

### **Curriculum Vitae of Dr. RAOUF O. LOUTFY**

#### **EDUCATION**

Diploma Business Administration, McGill University, 1976  
Ph.D. Physical Chemistry/Electrochemistry, University of Western Ontario, 1971  
M.Sc. Solid State, American University, 1966  
B.Sc. Applied Chemistry, Cairo University, 1964

#### **PROFESSIONAL EXPERIENCE**

**1999 – Present COO, FIC Corporation, New York, NY**  
Responsible for technical direction of this joint venture between MER, Mitsubishi Corporation, and RCT Corporation for the commercialization of Fullerene and Fullerene based materials.

**1985 – Present President, MER Corporation, Tucson, Arizona**  
Responsible for developing advanced programs and technologies, managing research and development group, contract administration and financial responsibilities. Produced SiC whiskers, SiC Fibers, ceramic-ceramic composites, advanced lithium-ion battery technology, low cost bipolar plates for fuel cell. Lead a group to develop scale-up production of fullerenes, and its applications development.

**1986 - 1988 President, Keramont Research Corp., Tucson. AZ**  
Responsible for building the infrastructure (equipment, personnel, and projects) for advanced materials research organization with emphasis on electronic ceramics, aluminum nitride substrates reinforcements ( $\text{SiC}_w$ ,  $\text{SiC}_f$  and  $\text{TiB}_{2w}$ ) and composites (ceramic-ceramic, and metal-ceramic. and intermetallic ceramic).

**1981 - 1985 Research Advisor, ARCO Metals/ARCO Chemical Company, Tucson. AZ**  
Major responsibility in the development of advanced technology to produce primary light metals and advanced materials, and to provide company-wide support in area of expertise. These efforts result, amongst others, in a commercial plant for the production of high purity alumina.

**1977–1981 Group Leader, Chemical Eng. Div., Argonne National Laboratory, IL**  
The major responsibility, as a group leader of the Electrolytic Technology Research Group, was to develop and implement a plan for the electrochemical technology for energy and resource saving. This was achieved by supporting and conducting research and development to improve industrial processes and identifying and developing new concepts of low energy alternative technologies. A second responsibility is the technical management of contracts in the electrolytic area and contracts for developing batteries for load-leveling applications.

**1972-1977 Group Leader, Noranda Research Center, Pointe Claire, Quebec**  
In charge of developing advanced pyro metallurgical and electrometallurgical processes to improve plant operations and profitability. Those efforts led to the development of DSA anodes for metal winning, and a new oxidant for zinc purification process.

**AWARDS**

<u>TITLE OF AWARD</u>	<u>DATE</u>	<u>PRESENTED BY</u>	<u>REASON FOR AWARD</u>
Industrial R&D 100	1990	R&D Magazine	Development of SiC Fibers
Corporate Entrepreneur of the year	1990	AIN	Most Number of Contract Won in Arizona
Industrial R&D 100	1991	R&D Magazine	Development of SiC Whisker
Product of the Year	1991	AIN	Fullerenes Production
Industrial R&D 100	1996	R&D Magazine	PDS Powder
Tibbetts Award	1998	SBA	SBIR Commercialization
Tibbetts Award	2001	SBA	Commercialization of Fullerene

**PUBLICATIONS AND PATENTS**

- 24 U.S. patents (12 have been in the last 10 years)
- 12 patent disclosures
- 90 articles published
- A chapter on Hydrogenated Fullerene in the Encyclopedia of Technology
- 2 books, both on Fullerenes:

D. Koruga, S. Hameroff, J. Withers, R.O. Loutfy, and M. Sundaresan  
 "Fullerene, C60: History, Physics, Nanobiology, and Nanotechnology"  
 Elsevier Science Publishing Co. New York, NY 1993.

"Perspectives of Fullerene Nanotechnology", edited by Eiji Osawa, Kluwer  
 Academic Publishers, February 2001.

The specific Chapters are:

1. R.O.Loutfy, A. Moravsky, A. Franco, and E.Veksler "Physical Hydrogen Storage on Nanotubes and Nanocarbon Materials"
2. R.O.Loutfy, S.Katagiri "Fullerene Materials for Lithium-ion Battery Applications"
3. R.O. Loutfy, S. Hossain, A. Moravsky and M. Saleh "Nanotubes as Anode Material for Lithium-ion Batteries"
4. Raouf O. Loutfy and Eugene M. Wexler "ABLATIVE AND FLAME-RETARDANT PROPERTIES OF FULLERENES"
5. R.O. LOUTFYJ. C. WITHERS, AND M. ABDELKADER "Development of Carbon Nanotube - Polymer Composites"
6. Raouf O. Loutfy, J.C.Withers and Stevan T. Dimitrijevic "USE OF FULLERENES AND CARBON NANOTUBES FOR FABRICATION OF EFFICIENT ELECTRON FIELD EMITTERS"
7. Raouf O. Loutfy, Eugene Wexler, and Weijiong Li "UNIQUE FULLERENE-BASED HIGHLY MICROPOROUS CARBONS FOR GAS STORAGE"
8. Raouf O. Loutfy, Timothy P. Lowe, Alexander P. Moravsky, and S. Katagiri "Commercial Production of Fullerenes and Carbon Nanotubes"
9. Raouf O. Loutfy, Eugene Wexler "GAS PHASE HYDROGENATION OF FULLERENES"

10. Raouf O. Loutfy, Eugene Weksler "HYDROGENATION OF ALKALI METAL - DOPED FULLERENES"
11. R.O.Loutfy, and M.Hecht "Aligned Carbon-Nanotubes for Sensor Applications"
12. Raouf O. Loutfy, Eugene Weksler "ADVANCED THERMAL PROTECTION COATING USING FULLERENES"

#### **Other Fullerenes Publications:**

S. Seraphin, J.C. Withers, R.O.Loutfy, et al, "TEM Studies of Nanotubes and Graphite Particles", Symposium of the Arizona Fullerene Consortium, November 6, 1992.

S. Seraphin, J.C. Withers, et al, "TEM Study of Carbon Nanotubes Produced by Various Processing Conditions," to be presented a John M. Crowley Symposium, Arizona State University, January 5-8, 1993.

S. Seraphin, J.Jiao, D. Zhou, J.C. Withers, R.O. Loutfy, "Effect of Processing Conditions on the Morphology and Yield of Carbon Nanotubes," *Carbon*, Vol 31, No 5, 685 (1993).

S. Seraphin, J. Jiao, D. Zhou, J.C. Withers, R.O. Loutfy, "Yttrium Carbide in Nanotubes," *Nature*, Vol. 362, April 8, 503 (1993).

J.C. Withers, R.O. Loutfy, K.Y. Donaldson, D.P. Hasselman, "Thermal Diffusivity/Conductivity of Compacts of C<sub>60</sub> Buckminsterfullerene and a C<sub>60</sub>/C<sub>70</sub> Mixture," *J.Am. Cerm. Soc.*, Vol 76, No 3, 754 (1993).

D. Koruga, J.S. Kustic, M. Trifunovic, S. Jankovic, S. Hameroff, J.C. Withers, R.O. Loutfy, "Imaging Fullerene C<sub>60</sub> with Atomic Resolution Using a Scanning Tunneling Microscopy," *J. of Full. Science & Technology*, Vol 1, 93 (1993).

R.O. Loutfy, J.C. Withers, "Fullerene & Electrochemical Hydrogen Storage" abstract 2145, Electrochem. Soc. Mtg, Hawaii, (1993).

J.C. Withers, R.O. Loutfy, "Production Process for Fullerenes, Review" Abstract #2127 Electrochem. Soc. Meeting, Hawaii (1993).

T. Yadav, S. Seraphin, D. Zhou, J.C. Withers, R.O. Loutfy, "Catalytic Growth of Buckyonions," in preparation

S. Supapan, D. Zhou, J.Jiao, M. Minke, S. Wang, T. Yadav, J.C. Withers, R.O. Loutfy, "The Effect of Pt, Pd and Ni on the Synthesis of Carbon Clusters," in preparation

J.C. Withers, C. Pan, R.O. Loutfy, "Fullerene Price: How Low Will They Be?" Electrochemical Society Mtg., San Francisco, Abstract 1216, (1994)

#### **Contracts**

DOE contract #DE-FG02-92ER81272, "The Development of a Process to Synthesis Tubular Fullerenes", Phase II, July 92

DOE contract #DE-FG02-91ER81095, "Novel C<sub>60</sub> Electrodes for Advanced Electrochemical Sensors", Phase I, Sept 91.

ARMY contract #DAAH04-93-C-0004, "Novel Materials for Hydrogen Supplies and Storage for Fuel Cells, Phase I, Jan 93.

ARMY contract #DASG60-93-C-0003, "Use of High Energy Lasers for Materials Synthesis", Phase I, Nov 92.

NASA contract #NAS5-32430 Goddard, "A Novel Negative Hydride Electrode for Ni-Metal Hydride Batteries", Phase I, Dec 92.

Exhibit #2



US006077401A

United States Patent [19]

Fields et al.

[11] Patent Number: 6,077,401

[45] Date of Patent: Jun. 20, 2000

[54] PRODUCTION OF FULLERENES USING  
CONCENTRATED SOLAR FLUX

[75] Inventors: Clark L. Fields, Greeley; John Roland  
Pitts; David E. Kling, both of  
Lakewood; Mary Jane Hale, Golden;  
Carl E. Bingham, Denver; Allan A.  
Lewandowski, Evergreen, all of Colo.

[73] Assignee: Midwest Research Institute, Kansas  
City, Mo.

[21] Appl. No.: 08/290,538

[22] Filed: Aug. 15, 1994

[51] Int. Cl.<sup>7</sup> C01B 31/02

[52] U.S. Cl. 204/157.47; 423/445 B;  
423/445 R

[58] Field of Search 423/445 B, 445 R;  
204/157.47

[56] References Cited

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5,316,636	5/1994	Bunshah et al.	423/445 B
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H. Kroto et al., "C<sub>60</sub>: Buckminster Fullerene", *Nature* 318,  
162 (1985).

Kratschmer, et al., *Nature*, 347 354 (1990).

J. Phys. chem. 97 (34), 8701 (1993), Chibante et al.  
Fields, C.L., et al. "Formation of Fullerenes in Highly  
Concentrated Solar Flux", *J. Phys. Chem.*, vol. 97 (Aug. 26,  
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IBM Technical Disclosure Bulletin, vol. 34, #4B, Sep. 1991,  
p. 222, Entitled "Method for Mass Producing Large  
'Fullerene' Type Carbon Clusters Using Localized Heating  
Sources".

Diederich, F., et al. *Science*, vol. 254, pp. 1768-1770 (Dec.  
20, 1991), Entitled "Fullerene Isomerism . . .".

McKinnon, J.T. "Calculated Equilibrium Yields of (60 From  
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pp. 8941-8944.

Primary Examiner—Michael Lewis

Assistant Examiner—Peter Di Mauro

Attorney, Agent, or Firm—Ken Richardson

[57] ABSTRACT

A method of producing soot containing high amounts of  
fullerenes comprising: providing a primary concentrator  
capable of impingement of a concentrated beam of sunlight  
onto a carbon source to cause vaporization of carbon and  
subsequent formation of fullerenes, or providing a solar  
furnace having a primary concentrator with a focal point that  
concentrates a solar beam of sunlight; providing a reflective  
secondary concentrator having an entrance aperture and an  
exit aperture at the focal point of the solar furnace; providing  
a carbon source at the exit aperture of the secondary con-  
centrator; supplying an inert gas over the carbon source to  
keep the secondary concentrator free from vaporized carbon;  
and impinging a concentrated beam of sunlight from the  
secondary concentrator on the carbon source to vaporize the  
carbon source into a soot containing high amounts of  
fullerenes.

20 Claims, 6 Drawing Sheets

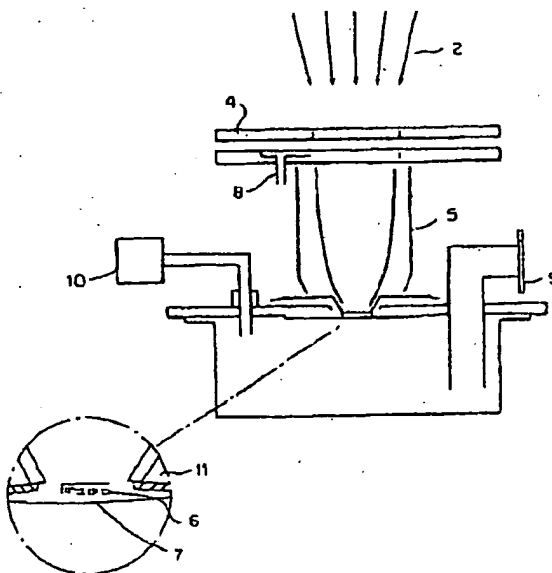


Exhibit 2

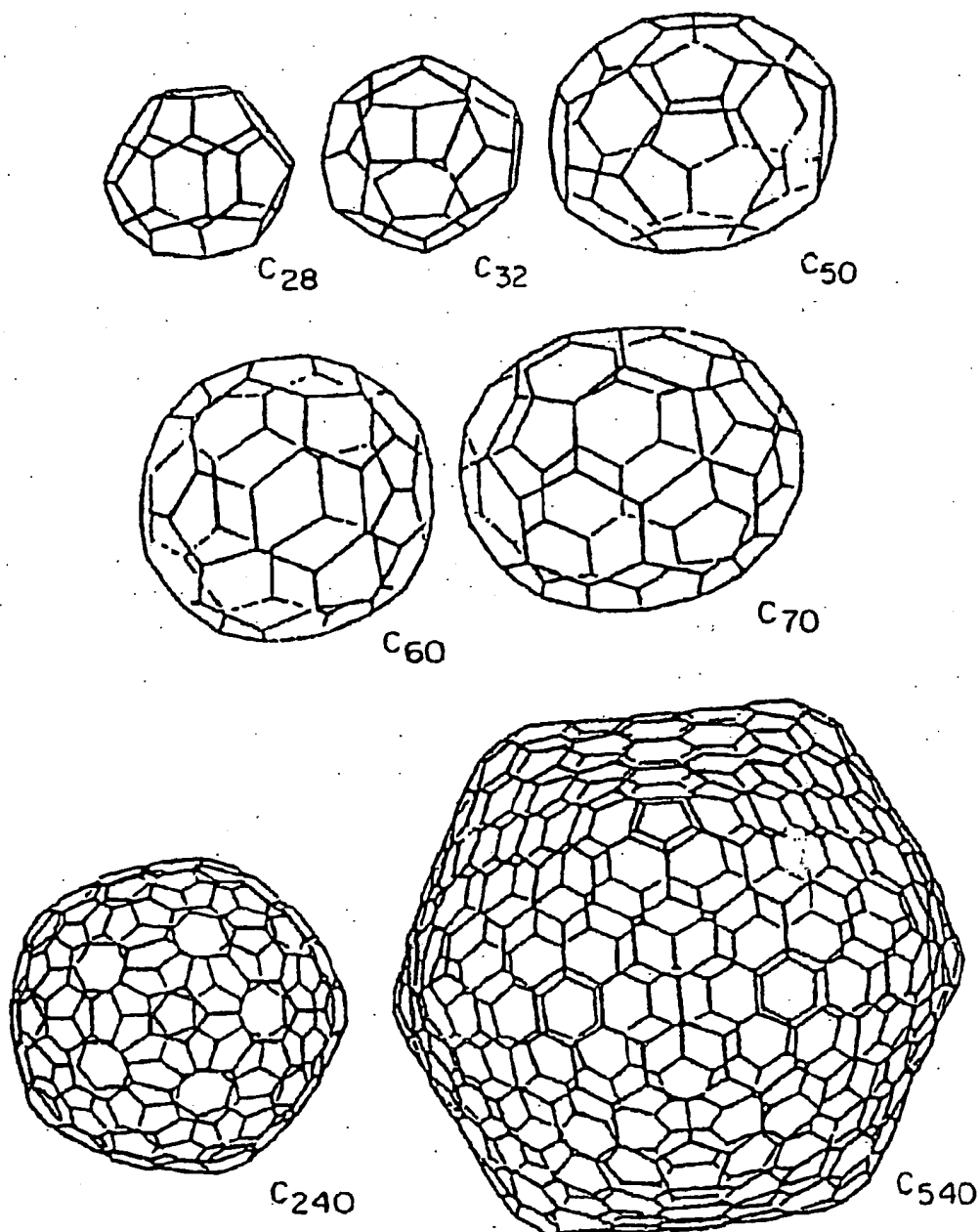
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FIG. 1



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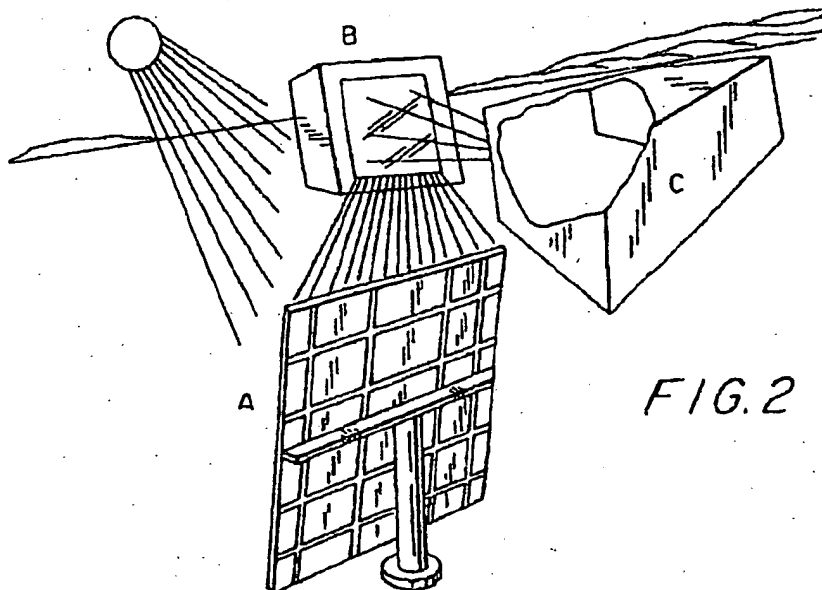


FIG. 2

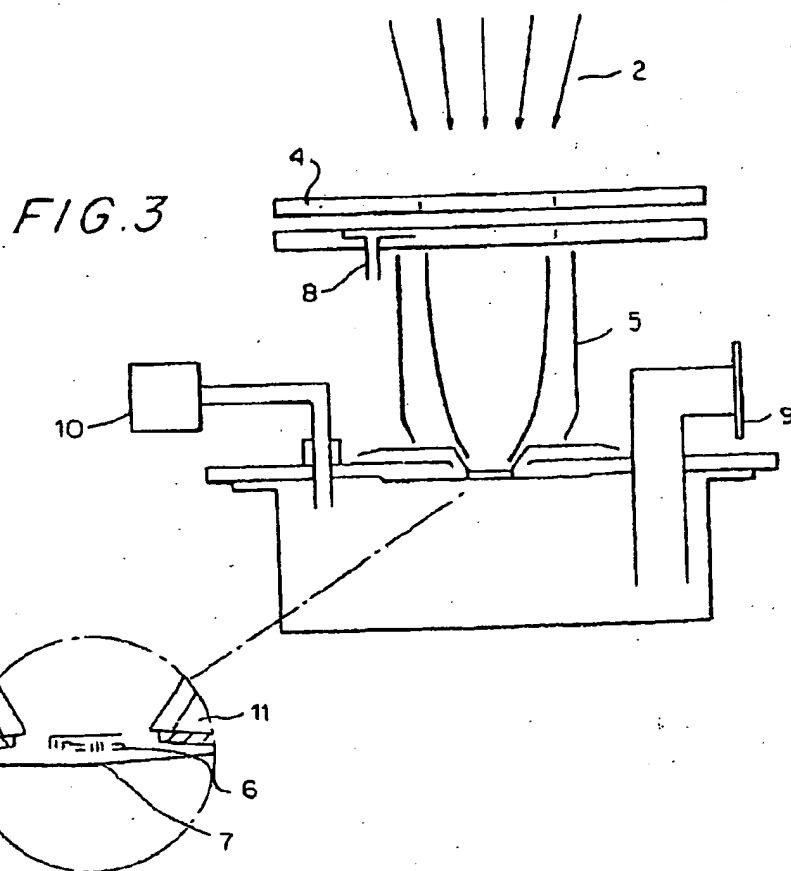


FIG. 3



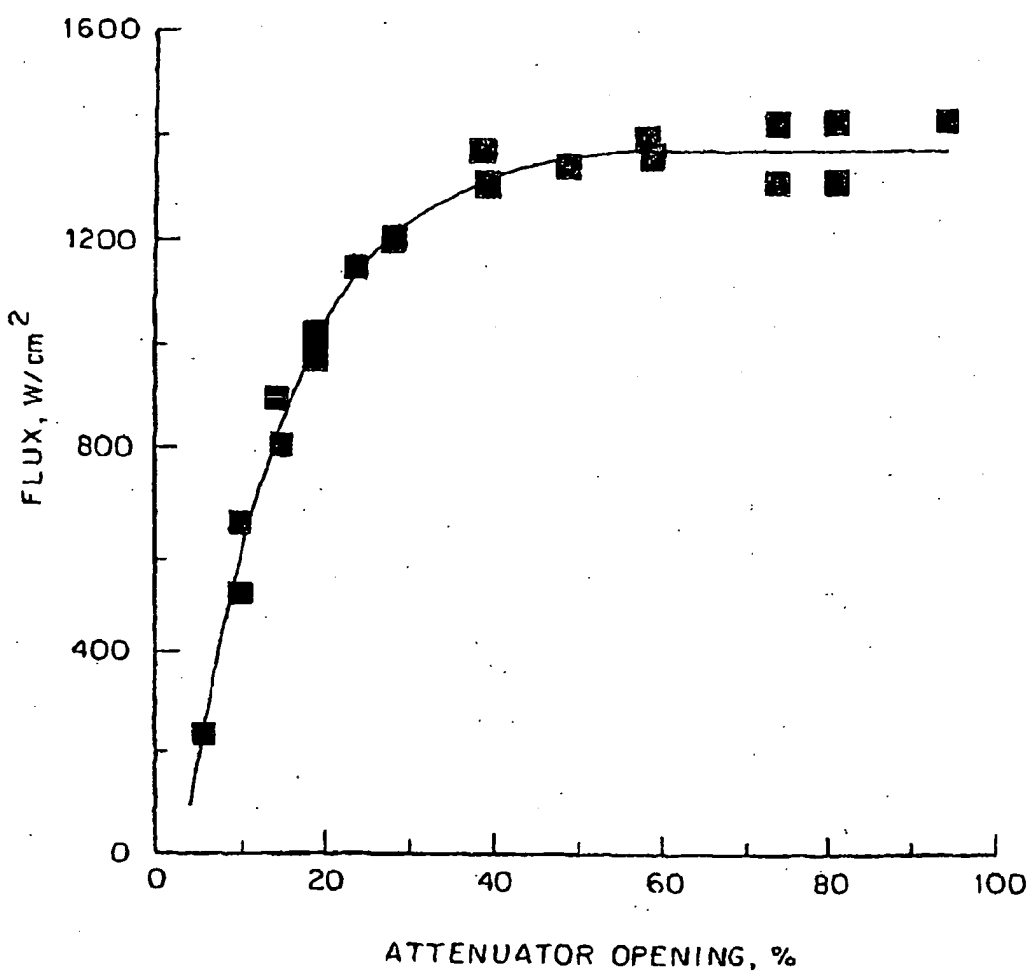
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FIG. 4



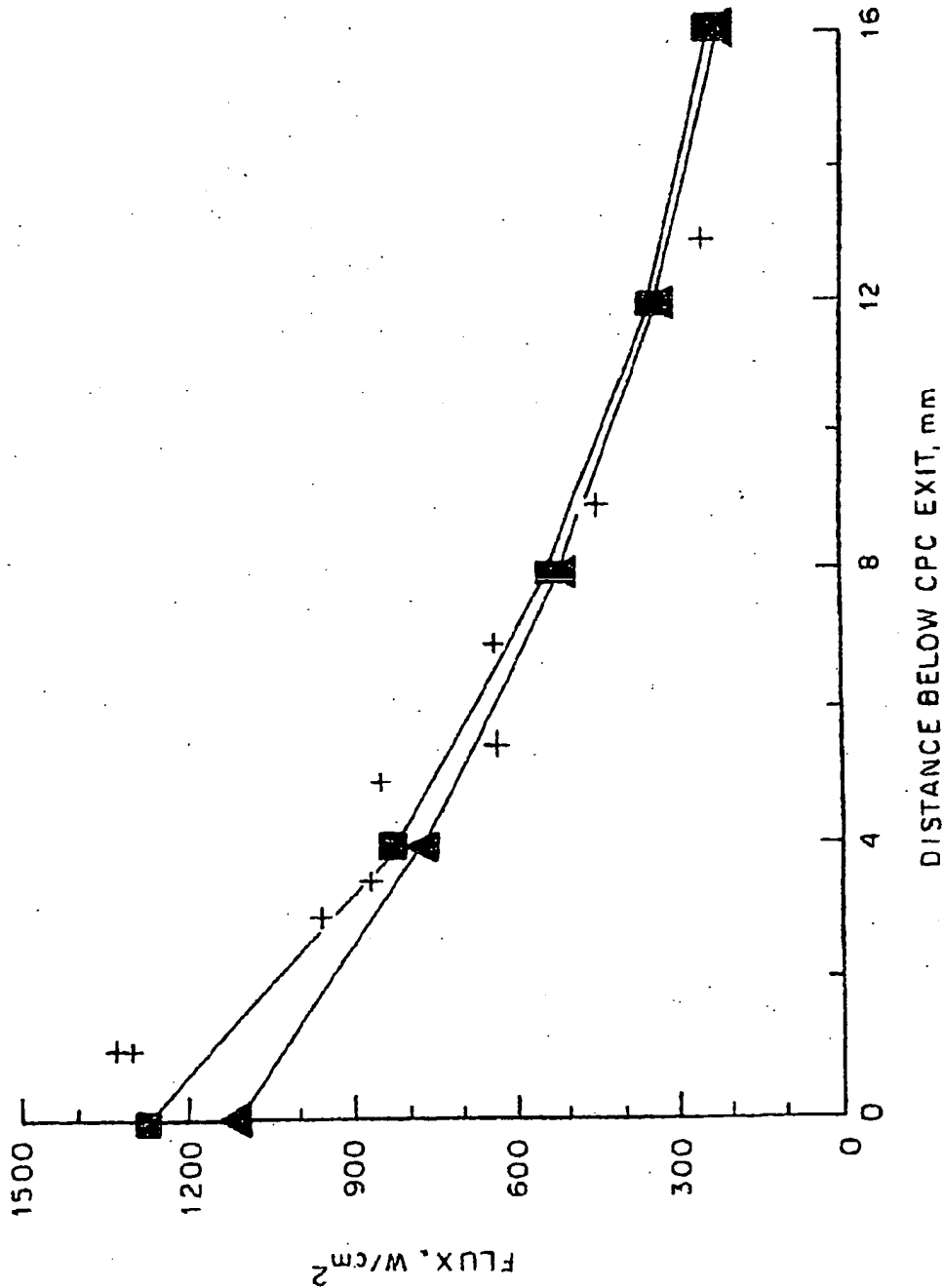
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FIG. 5



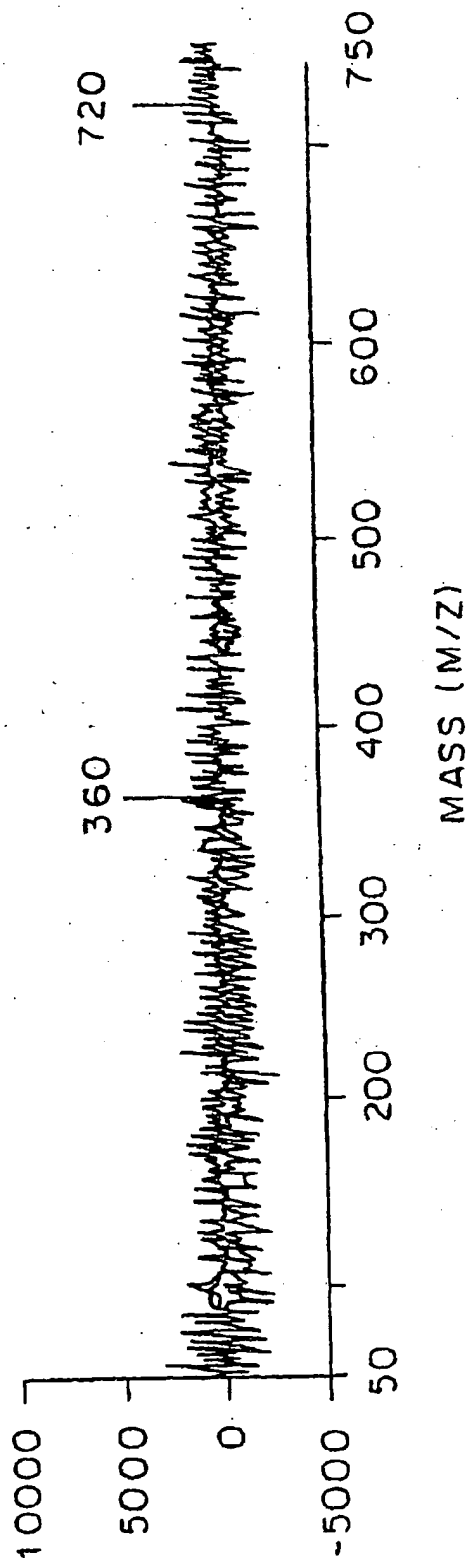
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FIG. 6



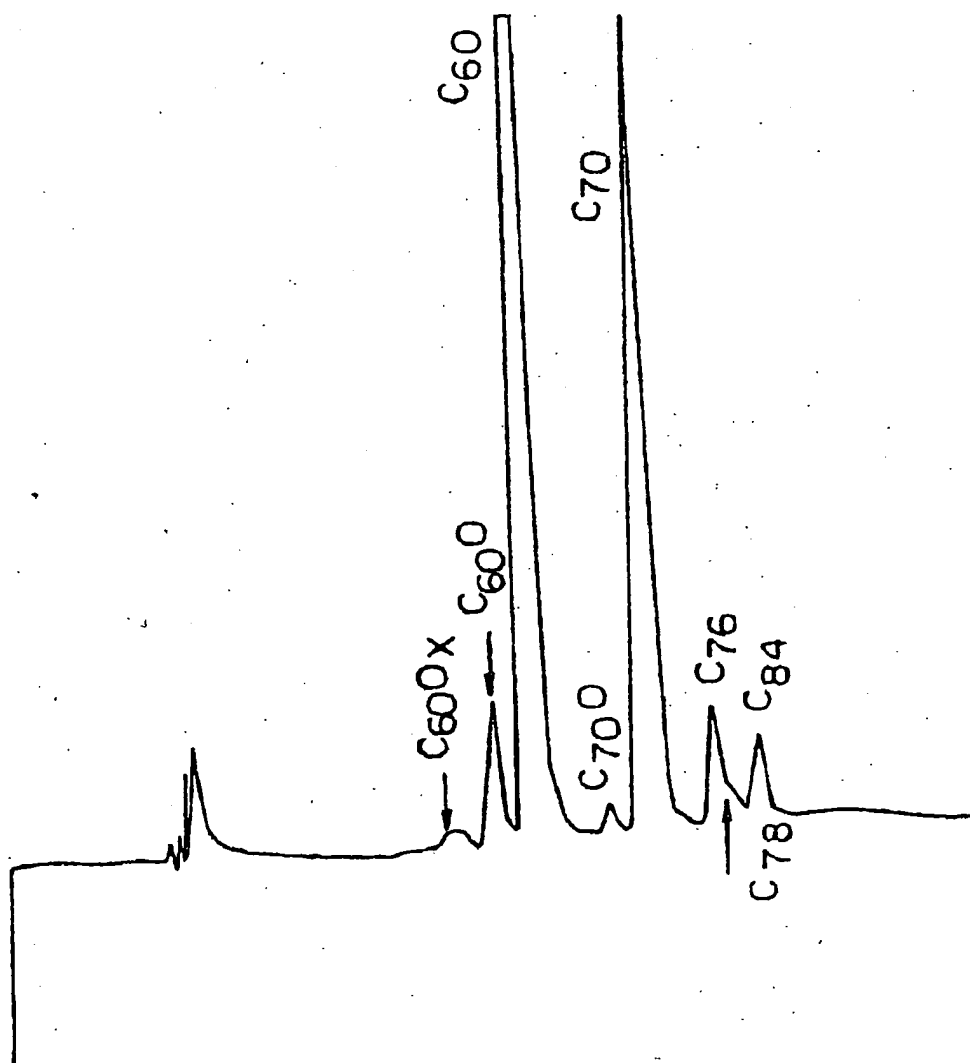
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FIG. 7



6,077,401

1

## PRODUCTION OF FULLERENES USING CONCENTRATED SOLAR FLUX

The United States Government has rights in this invention under Contract No. DE-AC02-83CH10093 between the United States Department of Energy and the National Renewable Energy Laboratory, a Division of the Midwest Research Institute.

### BACKGROUND OF THE INVENTION

#### 1. Field of the Invention

The present invention relates generally to a process for delivering highly concentrated solar radiation to a material surface to evaporate the material so that it condenses into caged molecules.

More specifically, the invention is directed to a method for producing fullerenes by: providing a solar furnace having a focal point wherein the solar furnace concentrates sunlight; providing a reflective secondary concentrator having an entrance aperture and an exit aperture at the focal point of the solar furnace; providing graphite at the exit aperture of the secondary concentrator; flowing argon gas over the graphite to keep the secondary concentrator free from vaporized carbon; and impinging the concentrated sunlight from the secondary concentrator onto the graphite to vaporize the graphite into a soot containing high amounts of fullerenes.

#### 2. Description of the Prior Art

A short while ago, carbon was considered to exist in a limited number of forms such as diamond, graphite, glassy carbon, amorphous carbon, and a number of high-temperature species that existed in the vapor phase above 2,000° C. However, in 1984, mass spectrometry experiments revealed that carbon could possibly exist in a number of other forms ranging from  $C_{30}$  to  $C_{100}$ .

Approximately one year subsequent to 1984, the unique stability of molecular allotropic forms such as  $C_{60}$  and  $C_{70}$  was demonstrated [H. W. Kroto et al.; *Nature* 318, 162 (1985)]. These events led to the discovery of a whole new set of carbon-based substances known as fullerenes. Fullerenes are composed of closed polyhedra or tubes produced by carbon atoms linking together to form hexagons and pentagons as shown in FIG. 1.

The configuration of carbon atoms in fullerenes provides properties that have captured the interest of chemists, physicists, materials scientists, and medical researchers, as fullerenes have been shown to crystallize to form interesting solids and to polymerize in several ways to form new polymers. Also, metal atoms can be placed inside the fullerene cage to form encapsulated systems (i.e.  $UC_{28}$ ,  $LaC_{82}$ , etc.), or outside the cage to form catalysts.

The fullerene cage can be reacted with other substances in a number of ways to form new molecules of interest.

Tubules of fullerenes have caught increasing interest as fibers, nanowires, and encapsulants. Fullerenes may also be doped to form electronic materials or reacted to form superconductors.

All of these applications have been discovered since the first macroscopic amounts of the most common fullerene,  $C_{60}$ , were isolated in 1990 [Kratschmer, et al., *Nature* 347, 354 (1990)].

Much of the work on fullerenes was performed using small amounts of material since synthetic approaches to these forms of carbon yielded limited quantities of material. The major drawback to the commercialization of some of the applications mentioned is due to the lack of a large-scaled method for producing and isolating fullerenes.

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Synthetic production of fullerenes was first provided using vaporization of graphite in an expanding helium atmosphere [H. W. Kroto, et al., *Nature* 318, 162 (1985)]. In this method, a Q-switched Nd:YAG laser is focused onto a rotating disc of graphite, whereupon carbon is evaporated or ablated into a high density helium flow. Clusters of soot form and are detected using a time-of-flight mass spectrometer. However, this method of production is sufficient to form only a few micrograms of fullerenes per day, which is only enough for certain limited research purposes.

A more useful method of synthesizing fullerene containing soot is the contact-arc method [Kratschmer, et al., *Nature* 347, 354 (1990)]. In this method, lightly contacting graphite electrodes are heated electrically by an alternating-current arc welder in an atmosphere of helium at a pressure of about 100 to about 200 torr. the graphite heats to evaporation at the contact and produces soot containing fullerenes. The soot condenses upon cool walls of a chamber, and is scraped off after a run that consumes the electrodes. Fullerenes are extracted from the soot by a solvent, such as toluene or benzene. This method is capable of producing a few tens of milligrams of fullerenes per run. While the apparatus used can be run in parallel so that the process is capable of producing several grams of fullerenes per day, the process is encumbered by scaling problems. For example, as the diameter of the rods are increased and the current supplied to the rods is increased to increase the amount of graphite evaporated per unit time, the yield of fullerenes decreases.

Rods that are  $\frac{1}{8}$ " in diameter are capable of producing maximum yields of about 30%, while rods that are  $\frac{3}{4}$ " in diameter are capable of producing yields of around 15%, and rods that are  $\frac{1}{2}$ " in diameter only have yields that are no more than 7%. The linear decrease in yield with an increase in rod diameter is not understood, but a reasonable conjecture put forth by Chibaue, et al. [(*J. Phys. Chem.* 97(34), 8696 (1993))], is that the intense ultraviolet light in the plasma region of the arc may destroy fullerenes before they can exit that region.

Howard, et al. in *Nature* 352,139 (1991) discloses a third method of producing fullerenes. This method entails burning hydrocarbon feeds in an oxygen deficient flame or sooty flame. Benzene is used as a hydrocarbon source, with an argon diluted oxygen supply. In this method, it was found that soot yields are 0.2 to 12% of the carbon feed, and this gives a maximum yield of fullerenes of 0.3% of the carbon feed. This synthesis process is too costly to compete with the contact-arc process.

U.S. Pat. No. 4,874,596 discloses a method of changing the structure of a solid material in the form of carbon by converting the carbon to one or more other forms including diamond by the intense heat and shock wave force generated and transmitted through the material by intense radiation. This process does not provide caged molecular forms (fullerenes) nor does it encroach upon the physical concepts required for their production.

The three prior art methods of producing fullerenes, namely, laser ablation of graphite targets, the carbon arc process (also called the contact-arc process) and the process whereby soot produced by an oxygen deficient flame is utilized are encumbered by: the small capability of producing only milligram quantities of fullerenes at most; loss of efficiency as the electrode diameter is increased; and the high expense-low yields of soot from benzene (about 0.5%) that result in  $C_{60}$  costs of at least about \$100.00/g.

Accordingly, there is a need extant in the art of producing fullerenes to provide a method for producing fullerenes that

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is greater than the milligram quantities presently available through current technology, by providing higher percentages of soot containing higher percentages of fullerenes, at lower cost.

### SUMMARY OF THE INVENTION

Accordingly, one object of the invention is to provide a method for producing fullerenes in quantities greater than the few milligrams of fullerenes produced per day by the laser ablation of graphite technique method.

A further object of the invention is to provide a method for producing fullerenes in greater quantities than the gram quantities of material produced per day per unit, without encountering loss of efficiency in production with increases in the electrode diameters using the carbon-arc process (also called the contact-arc process).

A yet further object of the invention is provide a method for producing fullerenes in which the amount of soot is greater in quantity than that which is produced by the oxygen deficient flame process, so that the amount of soot and the quantity of fullerenes contained therein is produced in greater yield.

A still further object of the invention is to provide a method for producing higher yields of fullerenes having a particular molecular weight in soot.

In general, the process of the invention is accomplished by delivering highly concentrated solar radiation onto a graphite target from a focal point of a solar furnace by: providing a solar furnace having a primary concentrator with a focal point that concentrates a solar beam; providing a reflective secondary concentrator having an entrance aperture and an exit aperture at the focal point of the solar furnace; providing graphite at the exit aperture of the secondary concentrator; flowing argon gas over the graphite to keep the secondary concentrator clean of vaporized carbon; and impinging the concentrated sunlight from the secondary concentrator onto the graphite to vaporize the graphite into fullerenes containing soot.

### BRIEF DESCRIPTION OF THE DRAWING

FIG. 1 shows spherically shaped fullerene molecules containing from  $C_{28}$  to  $C_{540}$  atoms, located at vertices.

FIG. 2 shows the high-flux solar furnace in which a heliostat (A) tracks the sun and directs the sunlight into a bank of concentrating mirrors (B), after which a concentrator array focuses the sunlight to a focal plane (target plane) inside a bay area (C), that houses the target, control room, and control functions of the furnace.

FIG. 3 shows the apparatus in which the graphite pellet is placed in proximity to the secondary concentrator.

FIG. 4 shows flux measurements, wherein the measured peak flux is plotted as a function of attenuator opening that makes flux in a given concentration available at the exit of the secondary concentrator.

FIG. 5 shows flux measurements plotted as a function of different distances from the secondary concentrator exit with different numbers of concentrator facets uncovered.

FIG. 6 shows a graph of a mass spectrum of  $C_{60}$  in soot produced at the solar furnace, where a 1-mg sample of soot was heated to 600° C. in flowing helium (5 L/min.) at the entrance orifice of a molecular beam mass spectrometer.

FIG. 7 is a graph showing High Pressure Liquid Chromatography (HPLC) of the toluene soluble extract from soot produced in the solar furnace using the process of the invention.

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### DETAILED DESCRIPTION OF THE INVENTION

All fullerene production methods require a source of small, gas phase carbon clusters (1 to possibly 10 atoms). Two sources of these clusters exist. One method is the combustion of hydrocarbons such as benzene in an oxygen deficient flame to produce a soot along with other combustion products such as  $H_2O$ ,  $CO$ , and  $CO_2$ .

While the percentage of fullerenes in the soot may be relatively high, the yield of fullerenes compared to mass of hydrocarbon consumed is low.

The second general method for production of fullerenes uses an energy source to vaporize elemental carbon at temperatures above 3000° C., whereupon the vaporized carbon then condenses into carbon soot. The number of energy sources utilized to vaporize the carbon include lasers, plasmas, induction heating, and arcs struck between graphite rods.

While the arc method has proven to be the most useful, even this method yields only tenths of grams of fullerenes per hour, and none of the existing methods have been brought to large-scale production.

Fullerenes are spherically or tubularly shaped molecules of carbon consisting of from 28 to more than 240 carbon atoms. They are sometimes called "Buckyballs", "Buckytubes", or "Buckminsterfullerenes" to describe the cage-like structures of the molecules. The typical "Buckyball" is the  $C_{60}$  molecule, which is a truncated icosahedron typified by the soccer ball, with carbon atoms at the 60 vertices of the pentagons and hexagons that form a nearly spherical surface.

Spherical shaped fullerene molecules containing from 28 to 540 carbon atoms are shown in FIG. 1.

Alternately, tubules (sometimes called nanotubes) can be formed where single (or multiple layers) of graphitically bonded ( $sp^2$ ) sheets are rolled to form a tube, which is capped by a hemispherical fullerene molecule.

Tubules can have diameters as small as 1 nm and they may have aspect ratios ( $l/d$ ) as large as  $10^4$  to  $10^5$ . The calculated tensile strengths of such fibers is enormous.

The system for providing fullerenes of the invention process utilizes highly concentrated sunlight delivered to graphite targets, so that the graphite evaporates and produces fullerenes. The heating process occurs solely from the application of radiant energy from the sun, and no other conversion processes are required. Further, no other energy resources are required, and fullerenes are produced in an energy efficient method without the environmental liabilities associated with fossil or nuclear energy.

The primary apparatus used is a high flux solar furnace at South Table Mountain in Golden, Colo. FIG. 2 is a photograph inclusive of a cut-away of the facility showing a heliostat (A) that tracks the sun and directs the sunlight into a bank of concentrating mirrors (B). The concentrator array focuses the sunlight to a focal plane (target plane) inside of the experimental bay area (C) that houses the target, control room, and all control functions for the furnace. Vacuum chambers, pumps, and a gas supply system are used inside of the bay room to control the processing environment.

The environmentally controlled target chamber consists of a target zone enclosed with a silica or quartz window to admit the sun, a gas supply system, and a vacuum system that allows very clean environments to be maintained. Base pressure in the vacuum system is  $10^{-8}$  torr, and it is equipped with a residual gas analyzer (RGA).

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In order to provide fullerenes in accordance with the invention, a process chamber was designed to incorporate a secondary concentrator to boost the flux to useful levels for evaporating graphite. A schematic of this chamber is shown in FIG. 3, where solar radiation from the primary concentrator passes through a quartz window 4 and through a secondary concentrator 5 having an entrance aperture and an exit aperture at the focal point of the solar furnace. A graphite target 6, placed on a target support 7 at the exit aperture of the secondary concentrator, is vaporized to fullerene containing soot by impinging concentrated sunlight. The apparatus which produces the soot containing fullerenes is equipped with a gas inlet 8 stacked on top of and in close proximity to the secondary concentrator. Argon gas is passed through the inlet port and through the secondary concentrator in a manner so that it impinges on the graphite sample and is then removed by a vacuum pump (not shown) through vacuum port 9. A capacitance manometer 10 is used to measure the gas pressure during the process.

It is important to note that the solar beam is admitted through a high purity fused silica window from the primary concentrator bank. Thereafter, the secondary concentrator concentrates the light down so that the flux is increased by about a factor of 10. The highly concentrated solar beam then impinges upon a graphite target, which may be a rod, button or shaped target or a receptacle containing graphite powder. A tantalum shield 11 deflects radiation from the target and prevents overheating of the stainless steel chamber.

Flux measurements were conducted to ensure that the graphite sample temperature was in the range of vaporization. The results of the flux measurements are shown in FIGS. 4 and 5, where the data in these figures were normalized to a direct-normal irradiance of 950 W/m<sup>2</sup>, which was representative for the days the tests were conducted.

In FIG. 4, the measured peak flux is plotted as a function of the attenuator opening. With the attenuator fully opened, a flux of over 1,300 W/cm<sup>2</sup> (corresponding to a 13,000 concentration just under 15,000) was available at the exit of the secondary concentrator. The data in FIG. 4 was measured with all 25 of the primary concentrator facets uncovered. Similarly, all of the facets were uncovered in the production of fullerenes.

The flux is plotted as a function of different distances from the secondary concentrator exit in FIG. 5. These measurements were compared to the flux predicted by a Monte Carlo computer code, SOLFUR, written to explicitly model the various optical components of solar furnaces. By using an additional code specific to reflective secondary concentrators, the set of rays from SOLFUR was traced from the entrance plane of the secondary concentrator to a plane placed at various distances below the exit. The data measured in FIG. 5 was gathered with 19 of the primary concentrator facets uncovered.

The reaction chamber of the apparatus used to produce fullerenes in the invention process is disposed vertically, and is a vacuum sealed system with gas flow ports that permit the fullerene production to be conducted in an oxygen and water free environment. It is sealed on top with a quartz window using a Viton O-ring.

The gas inlet port and the secondary concentrator are disposed beneath the quartz window, and a needle valve outside of the reaction chamber regulates gas flow into the chamber.

The carbon sample sits at the exit aperture of the secondary concentrator, which is in the region of highest solar flux.

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The secondary concentrator and the horizontal surface just beneath it have internal cooling channels that are connected to cooling lines.

The bottom section of the reaction chamber is connected to a pumping system fitted with a mechanical pump, a turbo molecular pump, a butterfly valve, and a residual gas analyzer (RGA).

When the concentrated solar flux (approximately 1300 W/cm<sup>2</sup>) impinges on the top surface of the graphite or carbon sample, the sample vaporizes and carbon vapor condenses in a dry ice trap and on the chamber walls as soot.

Throughout the run, an inert gas (argon) was passed from the inlet port, through the secondary concentrator, over the carbon sample, and then out of the vessel through the vacuum pumping port. In addition to providing an oxygen-free environment, the flowing gas helps to cool the system and keeps the secondary concentrator clean by sweeping the carbon vapor away from its reflective surface.

Graphite was used as the carbon source and the samples were cut from a 0.75-cm thick slab of 99.999% purity graphite. Several 1-cm-diameter cylinders were cut from this slab. Unmodified and modified graphite cylinders were used in the runs. Modified graphite samples were modified by bevelling the top edge to form a conical shape or by cutting a small, hemispherical cavity into the top surface.

For each run, a graphite sample was placed at the exit of the secondary concentrator and the secondary concentrator was at the High Flux Solar Furnace's (HFSF's) focal point. After connecting the chamber to the vacuum system and the cooling lines, the reaction chamber was subjected to a pumping and purging process to clear it of air and water vapor. The chamber was then pumped down to 50 micorr, and backfilled with argon to 400 torr. This pumping followed by purging was repeated four times, after which the argon inlet valve was closed, and the vessel was pumped down to 5x10<sup>-3</sup> torr to ensure that it was not leaking.

The argon flow was adjusted to the proper level and the butterfly-valve setting was adjusted to bring the chamber to the desired pressure (either 50 torr or 100 torr). Once the chamber pressure was steady, the chamber was illuminated with the attenuated concentrated solar beam for approximately three minutes to "bake out" any impurities in or on the graphite sample. After this baking-out period, the sample was removed from the chamber, weighed, and replaced.

The above procedures were repeated beginning with pumping and purging and finishing with the chamber pressure adjusting. The graphite sample was then exposed to the concentrated beam for a period of from about 30 seconds to about two minutes. After exposure to the flux, the reaction chamber was opened and the soot was removed from the condensing surfaces with a small brush, bottled, and the graphite sample was reweighed.

As shown in the graph of FIG. 6, a mass spectrum of C<sub>60</sub> in soot was produced at the solar furnace, where a 1-mg sample of soot was heated to 600° C. in flowing helium (5 liters/minute) at the entrance orifice of a molecular beam mass spectrometer. In this graph, the vertical axis shows relative intensity and the horizontal axis shows the mass. The mass spectrum shows significant peaks at 360 amu and 720 amu for the doubly and singly charged C<sub>60</sub> molecules.

There were several variables in the operating conditions that had the potential of affecting the optimization of fullerene yield. To determine the influence of the different variables, a Plackett-Burman design of balanced incomplete blocks was used to design a set of runs.

The Plackett-Burman design is a two-level method ("plus" denoting high level and "minus" low level), of

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experimental design where as few as N experiments can be used to study the effect of as many as N-1 variables (Stowe et al., "Efficient Screenings of Process Variables", Industrial and Engineering Chemistry, 1966, pp. 36-40).

The variables are usually associated with an experimental parameter (i.e., gas pressure), but they can be dummy variables. Dummy variables are not associated with any experimental parameters, but instead add more experiments to the design and thereby refine the statistical calculations of variable effect and standard error.

According to the Plackett-Burman design, the effect of any one variable on the experimental result is determined by the difference between the average value of the results for the positive runs less the average value of the results for the negative runs, as per Equation 1.

$$E_{\text{variable}} = \frac{\sum R_{(+)}}{\text{No. of (+) runs}} - \frac{\sum R_{(-)}}{\text{No. of (-) runs}} \quad \text{Eqn. (1)}$$

where,

$E_{\text{variable}}$  = effect of variable on results (% fullerene yield),  
 $R_{(+)}$  = results from experiments with variable at positive value,

$R_{(-)}$  = results from experiments with variables at negative value.

The standard error in the effect is calculated with Equation 2.

$$SE_{\text{effect}} = \sqrt{\frac{\sum (E_{\text{dummy}})^2}{n}} \quad \text{Eqn. (2)}$$

where,

$SE_{\text{effect}}$  = standard error of any effect calculate with Equation 2,

$E_{\text{dummy}}$  = effect of dummy variable(s) on result,  
 $n$  = number of dummy variables.

In the operating runs, the effects of seven different variables were considered. According to the Plackett-Burman method, this required a set of eight runs. The variables and their high-and-low values are given in Table 1. Table 2 has the distribution of the high and low parameters throughout the set of eight runs.

TABLE 1

Variables and parameters used for the Plackett-Burman design		
Variable	High Parameter (+)	Low Parameter (-)
A = Solar flux	1300 W/cm <sup>2</sup>	1150 W/cm <sup>2</sup>
B = Position of sample	at secondary concentrator exit	2 mm below concentrator exit
C = Sample shape	cavity	cone
D = Length of exposure	2 min	1 min
E = Gas flow rate	4 slm	2 slm
F = Type of gas	He	Ar
G = Gas pressure	100 torr	50 torr

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TABLE 2

Plackett-Burman matrix for parameters							
Run	A	B	C	D	E	F	G
1	+	+	+	-	+	-	-
2	+	+	-	+	-	-	+
3	+	-	+	-	-	+	+
4	-	+	-	-	+	+	+
5	+	-	-	+	+	+	-
6	-	+	+	+	+	-	+
7	-	+	+	+	+	+	-
8	-	-	-	-	-	-	-

A series of preliminary runs were made to determine the high-low limits given in Table 1. For example, in one set of runs, it was observed that very little soot was produced for exposure times less than one minute. As the exposure time was lengthened beyond one minute, it was observed that the temperature of the apparatus became dangerously high after approximately two minutes. Therefore, the type of gas was kept constant throughout the set of eight runs and the variance "F" became a dummy variable. It appeared that the superior heat transfer characteristics of helium, as compared to argon, prevented the carbon sample from reaching a high enough temperature for vaporization.

Again, during the preliminary runs, mass spectrometry was used to confirm the existence of  $C_{60}$  in the collected soot, and FIG. 6 shows the mass spectrum of a 1 mg soot sample produced during preliminary runs.

The set of eight runs were performed under the conditions prescribed in Table 2.

Table 3 gives the mass of soot produced during each of the runs and the percentage of  $C_{60}$  in each soot sample.

TABLE 3

Masses of soot produced during two level experiments and corresponding fullerene yield		
Run	Soot Mass (mg)	% Yield of Fullerenes
1	7.0	3.2
2	7.6	12.8
3	7.0	3.2
4	0	NA
5	4.7	7.3
6	0	NA
7	12.6	13.9
8	0	NA

In Table 3, it is shown that only half of the runs resulted in appreciable soot production. Examination of the data reveals that all of the runs with the sample at the concentrator exit resulted in a measurable amount of soot, but no soot was produced in the runs where the sample was about 2 mm or more below the concentrator exit. Therefore, the graphite sample should be 2 mm or less below the secondary concentrator exit. This is an indication that the negative value for the sample positioning was too extreme.

Of the soot-producing runs, run 7 showed the most promising results, as this run resulted in the largest amount of soot and the highest yield. Although the difference in the percent yield between runs 7 and 2 appears to be within the margin of experimental error (13.9% vs. 12.8%), the difference in the amount of fullerenes produced is significant. Run 7 resulted in approximately 1.8 mg of fullerenes compared to just under 1 mg in run 2. Run 2 and run 7 were both



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performed with a high exposure length and a low gas flow rate, but run 7 was performed with a low flux parameter, a cavity-shaped sample, and a low chamber pressure.

During these runs, it was apparent that the cavity-shaped samples often showed much greater signs of vaporization activity than the conical-shaped samples. Sample shape is therefore the likely parameter responsible for the large amounts of soot (and hence greater fullerene production) in run 7.

FIG. 7 shows the retention-time plot from the HPLC analysis of soot run 7.

The effects of the different experimental variables and the associated standard error were calculated using Equations 1 and 2. These statistical results are given in Table 4.

TABLE 4

Relative effects of experimental variables and the standard error on percent fullerene yields.	
Variables	Effect
Position of sample (B)	9.3
Length of exposure (D)	4.05
Gas flow rate (E)	4.05
Solar flux (A)	1.3
Dummy (F)	1.3
Sample shape (C)	0.75
Chamber pressure (G)	0.75
Standard Error	1.3

The relatively large standard error in the effect calculations eliminates the significance of the effects calculated for three of the variables: the solar flux (A); the shape of the sample (C); and the chamber pressure (G). However, the calculated effects of the remaining three variables are outside of the standard error range.

It is apparent that the position of the sample relative to the secondary concentrator exit had the most significant effect.

The effects of the length of exposure and the gas flow rate calculated to be the same. The fact that no soot was ever produced when the carbon sample was in its low position increased the likelihood for two effects to calculate the same value. The fact that the four runs with low sample position resulted in no soot production lessened the impacts of these runs. The effects of the length of exposure and the gas flow rate were probably not equally significant to the fullerene yield.

It was noted that the effect of the rate of gas flow was actually a negative value; in other words, a low gas flow rate was more conducive to fullerene production.

It appears that the high gas flow cooled the graphite sample, and thereby inhibited vaporization.

Although the low sample position was too extreme to realize the full benefit of the Plackett-Burman design, the effect analysis was still beneficial. The calculated effects revealed that the carbon sample should be positioned at the concentrator exit, and that the lower gas flow rate and the longer exposure times were both conducive to high yields of fullerenes.

Percent yields for the soot samples were determined by toluene extractions from run 7 and matching their high-pressure liquid chromatography (HPLC) retention times with toluene extracts from commercially available soot samples. The yield analysis was performed on a Hewlett Packard Model 1050 HPLC Instrument using a reverse phase column (HP 5 $\mu$  silica, 300 Å pore size) with toluene/methanol (45:55 at 2 mL/min) as the eluant, and the results are shown in FIG. 7.

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While the preferred method of producing soot containing high amounts of fullerenes will utilize a solar furnace having a primary concentrator, followed by the use of a secondary concentrator having an entrance aperture and an exit aperture at the focal point of the solar furnace so as to affect impingement of a concentrated beam of sunlight from the secondary concentrator onto the carbon source to vaporize the carbon source into soot containing high amounts of fullerenes, it is to be understood that the use of a primary concentrator alone will suffice to provide producing soot containing high amounts of fullerenes if the concentrated beam of sunlight is sufficient to cause evaporation of carbon and subsequent formation of fullerenes. In the context of the invention, it has been found that, when a primary concentrator is focused to provide a concentrated solar beam having a flux of between about 250 W/cm<sup>2</sup> to about 2,000 W/cm<sup>2</sup>, the carbon source will vaporize into a soot containing high amounts of fullerenes.

As a result of the invention process, larger amounts of fullerenes can be economically prepared from soot obtained using concentrated solar flux as a supply of source material. The present invention therefore provides a highly economical and useful process for producing fullerenes.

While fullerenes of clusters of carbon atoms ranging from C<sub>20</sub>-C<sub>540</sub> range were produced, only small amounts of fullerenes having carbon atoms greater than the C<sub>60</sub> and C<sub>70</sub> (the most common forms of fullerenes) were produced.

In selecting the carbon source material for production of fullerenes in accordance with the invention process, diamond, graphite, graphite powder, glassy carbon, amorphous carbon, hydrocarbon feed and benzene will suffice; however, graphite is preferred. Graphite having a hemispherical cavity in its top surface is most preferred, as it provides greater amounts of soot containing higher amounts of fullerenes.

Any of the inert gases will suffice in the context of the invention for purposes of keeping the secondary concentrator free from vaporized carbon; however, argon is preferred as it has less of a tendency to prevent vaporization of the soot produced in the solar furnace.

The foregoing description is illustrative only of the principals of the invention. Further, since numerous modifications and changes will readily occur to those skilled in the art, it is not desired to limit the invention to the exact construction and operation shown and described, and accordingly, all suitable modifications and equivalence may be resorted to within the scope of the invention as defined by the claims which follow.

The embodiments of the invention in which an exclusive property or privilege is claimed are defined as follows.

What is claimed is:

1. A method of producing soot containing fullerenes solely from the application of radiant energy from the sun to a carbon source material comprising:

providing a solar furnace having a primary concentrator with a focal point that concentrates a solar beam of sunlight;

providing a reflective secondary concentrator having an entrance aperture and an exit aperture at the focal point of the solar furnace;

providing a carbon source material less than about 2 mm below the exit aperture of the secondary concentrator; supplying an inert gas over the carbon source material to keep the secondary concentrator free from vaporized carbon; and

impinging a concentrated beam of sunlight from the secondary concentrator onto the carbon source material

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less than about 2 mm below said exit aperture to vaporize the carbon source material into a soot containing fullerenes.

2. The method of claim 1, wherein the concentrated solar flux is between about 250 W/cm<sup>2</sup> to about 2,000 W/cm<sup>2</sup>.

3. The method of claim 1, wherein the fullerenes in the soot are selected from the group consisting of clusters of carbon atoms in the C<sub>28</sub>-C<sub>540</sub> range.

4. The method of claim 3, wherein the fullerenes in the soot are selected from a group consisting of clusters of carbon atoms in the C<sub>60</sub>-C<sub>70</sub> range.

5. The method of claim 3, wherein the solar furnace is vacuum-sealed and has a top section which includes a quartz window.

6. The process of claim 5, wherein the carbon source is selected from the group consisting of: diamond, graphite, amorphous carbon, and hydrocarbon feed.

7. The process of claim 6, wherein the carbon source is graphite.

8. The process of claim 7, wherein the inert gas is selected from the group consisting of helium and argon.

9. The process of claim 8, wherein the inert gas is argon.

10. The process of claim 9, wherein the concentrated solar flux from the primary concentrator has a 12 cm diameter and a peak flux of about 250 W/cm<sup>2</sup>.

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11. The process of claim 10, wherein the secondary concentrator has a concentrated solar flux range of between about 1300 W/cm<sup>2</sup> and about 2000 W/cm<sup>2</sup>.

12. The process of claim 11, wherein the graphite is in the form of a cylinder.

13. The process of claim 12, wherein the graphite is in a conically shaped form.

14. The process of claim 11, wherein the graphite has a hemispherical cavity in a top surface thereof.

15. The process of claim 11, wherein the graphite is exposed to the concentrated beam from the secondary concentrator for a period of from about 30 seconds to about 2 minutes.

16. The process of claim 15, wherein the fullerene is a C<sub>60</sub> fullerene.

17. The process of claim 15, wherein the fullerene is a C<sub>70</sub> fullerene.

18. The process of claim 15, wherein the fullerene is a C<sub>76</sub> fullerene.

19. The process of claim 15, wherein the fullerene is a C<sub>78</sub> fullerene.

20. The process of claim 15, wherein the fullerene is a C<sub>84</sub> fullerene.

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